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In-situ Monitoring of Dynamic Phenomena during Solidification and Phase Transformation Processing

July 25, 2012

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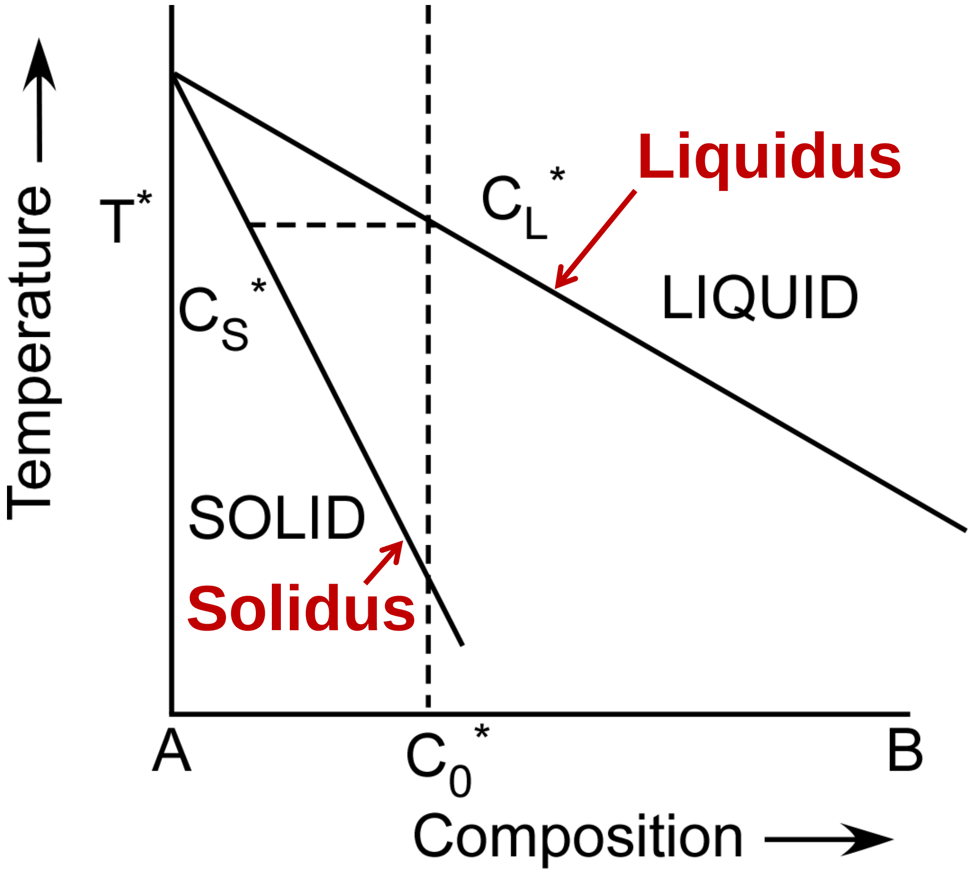
Purpose

- Directly observe phase transformations and microstructure evolution using proton (and synchrotron x-ray) radiography and tomography
- Constrain phase-field models for microstructure evolution
- Experimentally control microstructure evolution during processing to enable co-design
- Advance toward the MaRIE vision

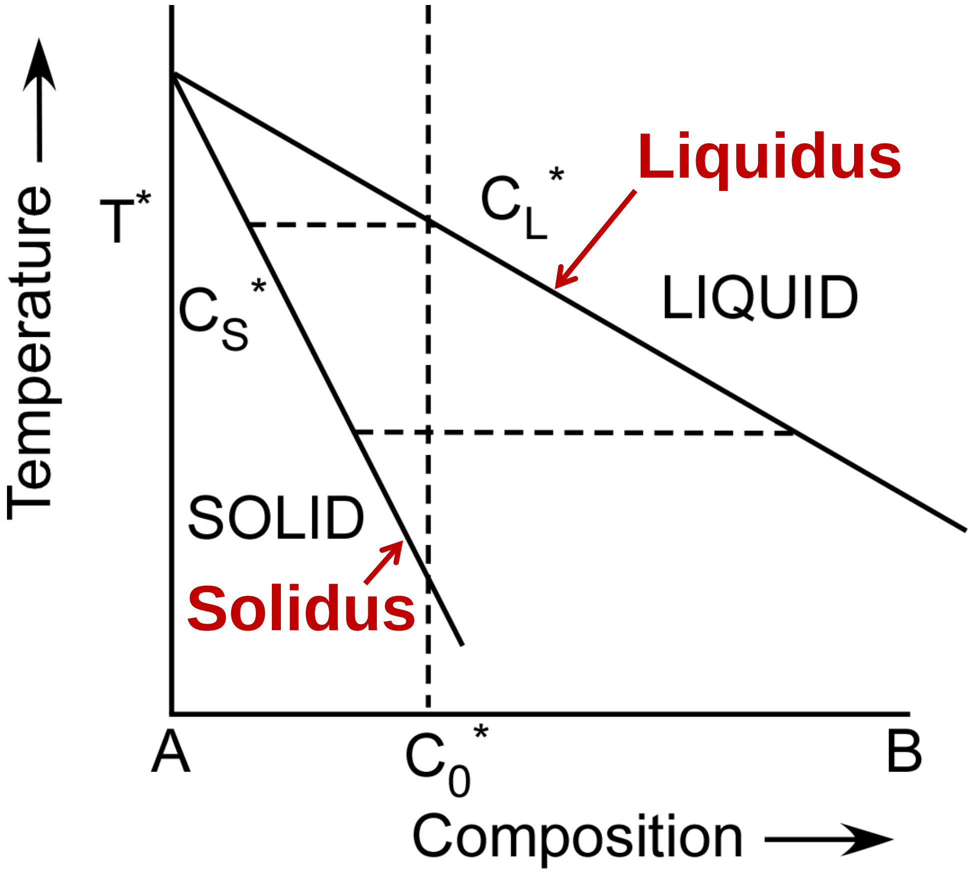
Understand microstructure evolution and chemical segregation during solidification → solid-state transformations in Pu-Ga

Phase Diagram for a Binary Alloy



- Pure metals solidify at a single T
- Alloys solidify over a range of T’s and compositions

Phase Diagram for a Binary Alloy



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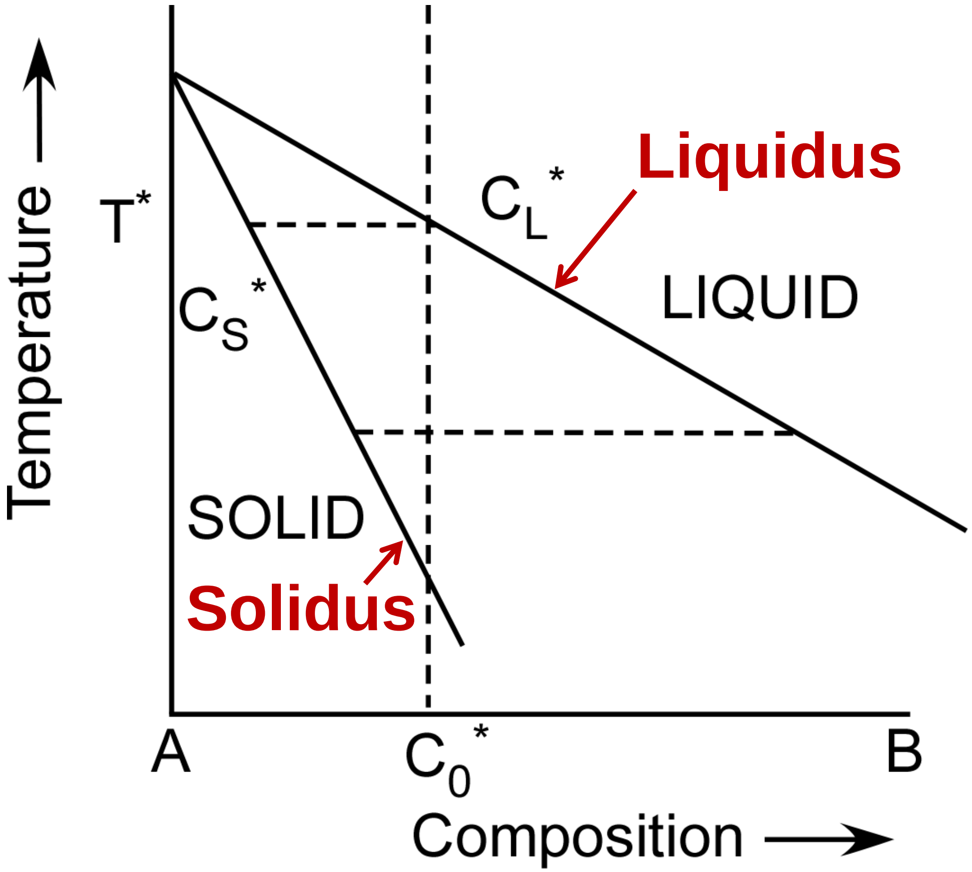
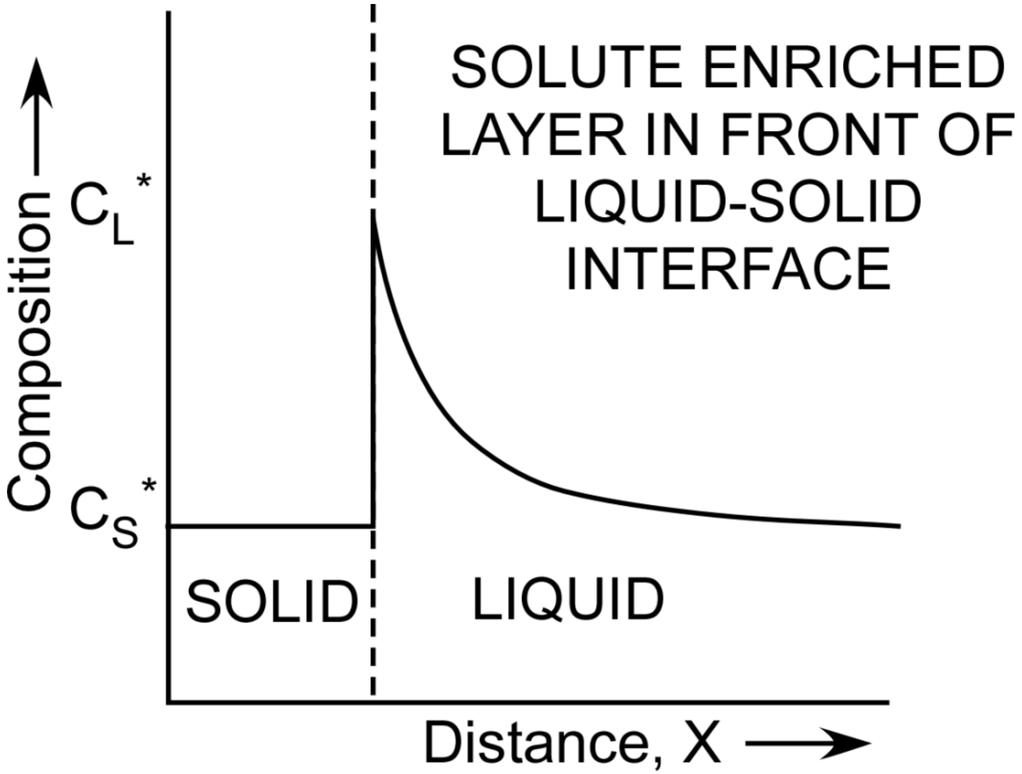
Phase Diagram for a Binary Alloy



- Pure metals solidify at a single T
- Alloys solidify over a range of T's and compositions
- Liquidus T decreases with increasing solute "B"
- Solid-state diffusion is slow
  → solute dumped into liquid
  → segregation or "coring"

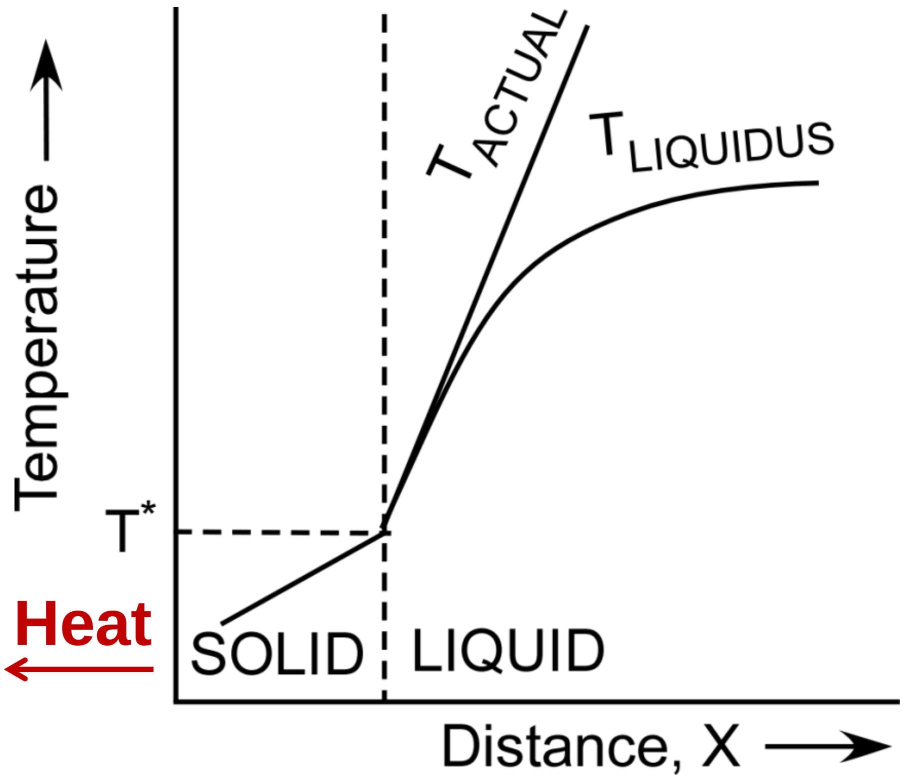
Composition Gradient Ahead of the Moving Interface



- Solute redistribution in the liquid occurs
- Composition gradient forms in the liquid
- Interface velocity (V) affects composition gradient

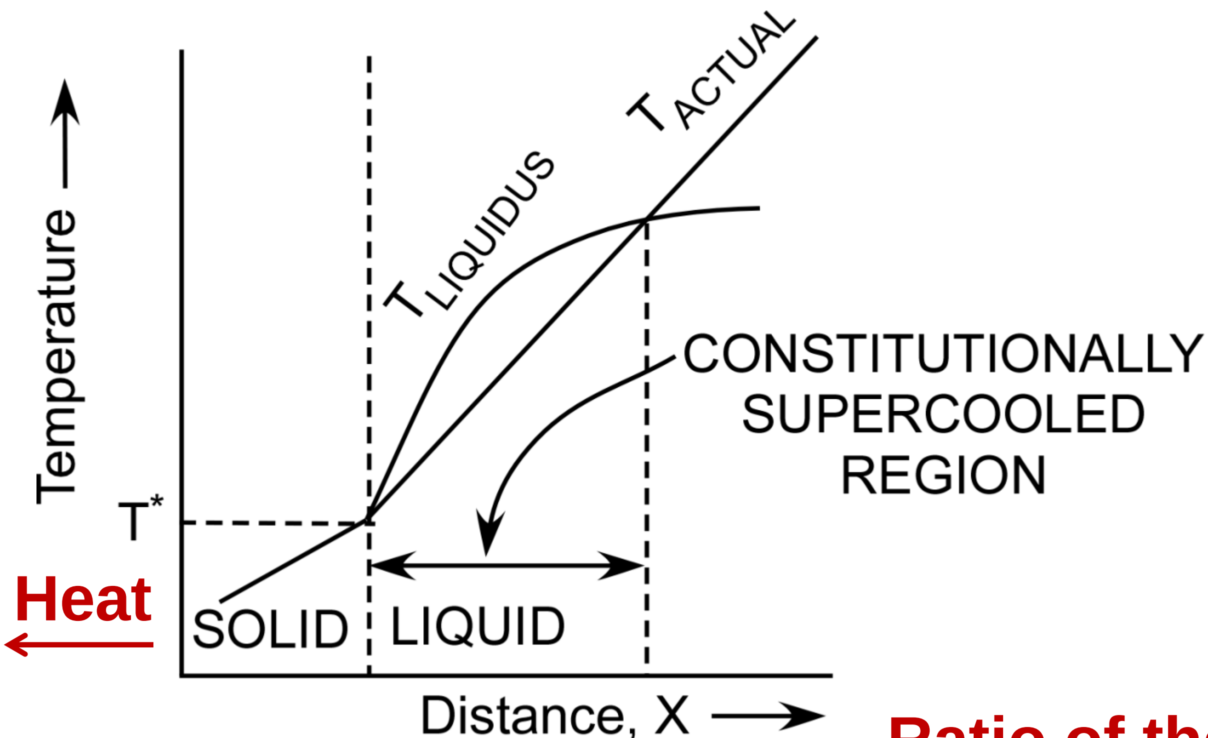
Composition gradient → $T_{LIQUIDUS}$ gradient

Interface Stability



- $T_{LIQUIDUS}(X)$ increases (solute decreases)
- Thermal gradient exists in the liquid and solid
- If $T_{ACTUAL} > T_{LIQUIDUS}$ (high thermal gradient G), plane front stable

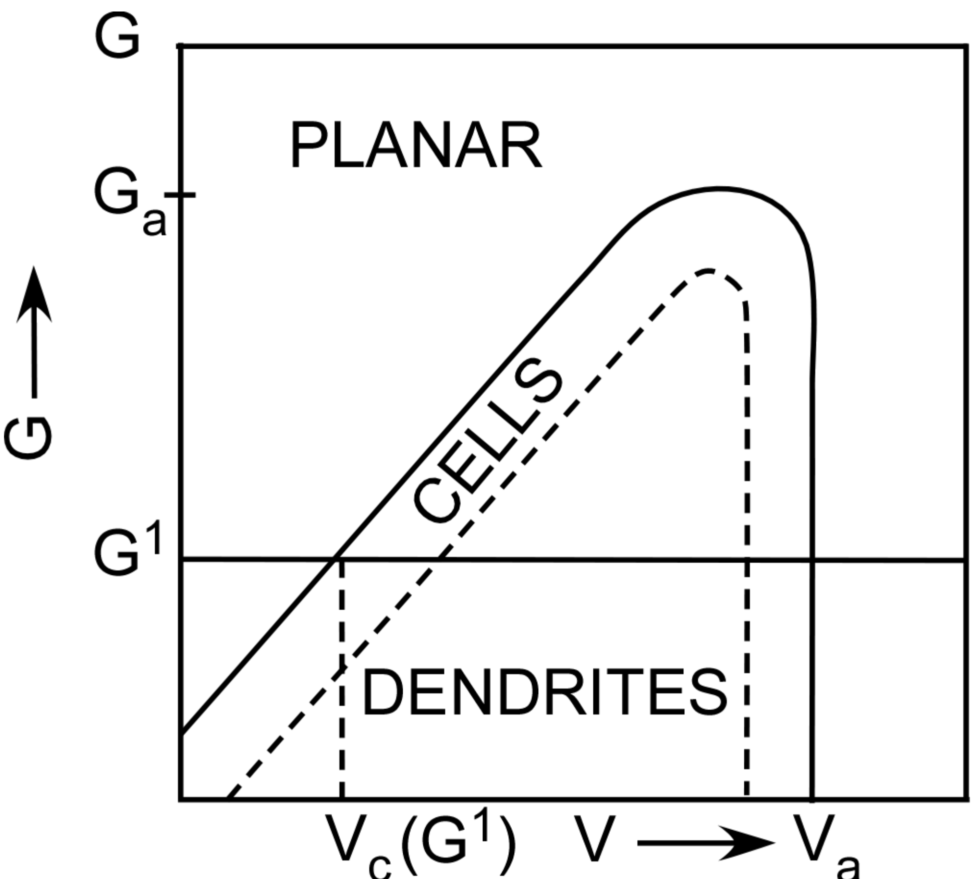
Interface Instability
- Constitutional Supercooling



- If $T_{ACTUAL} < T_{LIQUIDUS}$ (low thermal gradient G), then a supercooled region exists
- Interfacial perturbations will grow → unstable interface

Ratio of the thermal gradient G and the interface velocity V determines the interface stability

Thermal Gradient (G), Interface Velocity (V), and Interface Stability



- Experimentally control G and V (morphology)
- Experiment constrained phase field models → predict microstructure and segregation as a function of processing parameters

Direct, real-time experiment/model interaction → science-based solidification processing

Nonlinear Surface Instabilities are Universal

Transparent organic analogs of metallic systems: pattern-forming instabilities

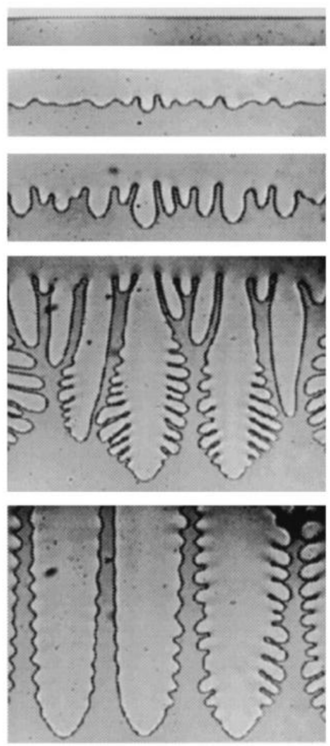
Stationary planar interface

Cellular pattern

Nonlinear instabilities

Coarsening phase

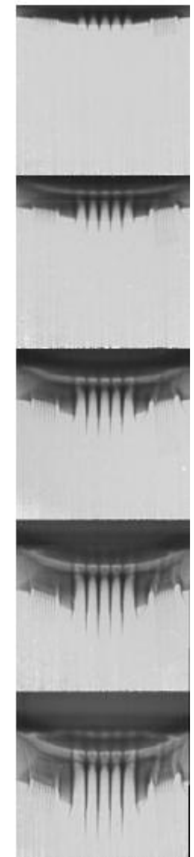
Steady-state dendritic array



Adapted from W. Losert et al., Proc Natl Acad Sci USA, 1998, 95:431

Increasing Time

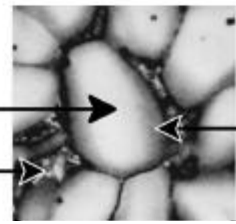
Richtmyer-Meshkov instability studies during shock (2009)

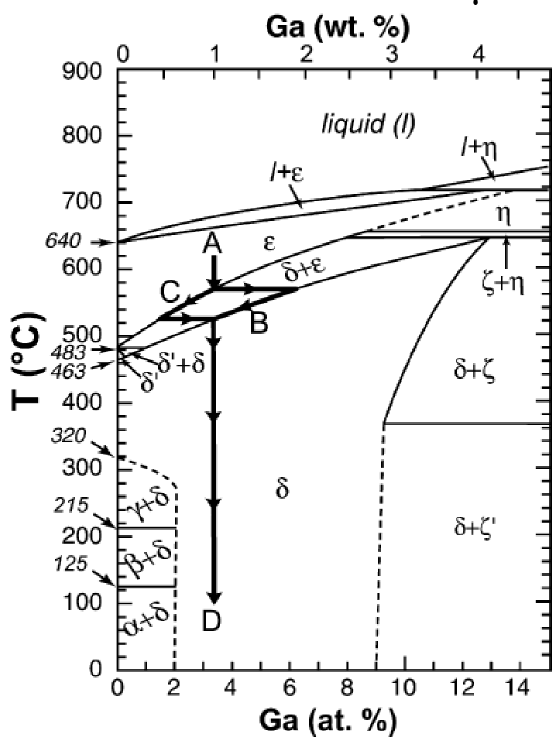


F.E. Merrill et al., Los Alamos National Laboratory, LA-UR 11-01518, 2011

W.T. Butler et al., Los Alamos National Laboratory, LA-UR 10-00734, 2010

“Solid-State Dendrites” in Pu-Ga?





Diffusion Rates:

- Liquid: ~10^{-6} cm^2/s (fast)
- ε-phase: 10^{-7} cm^2/s
- δ-phase: 10^{-10} cm^2/s

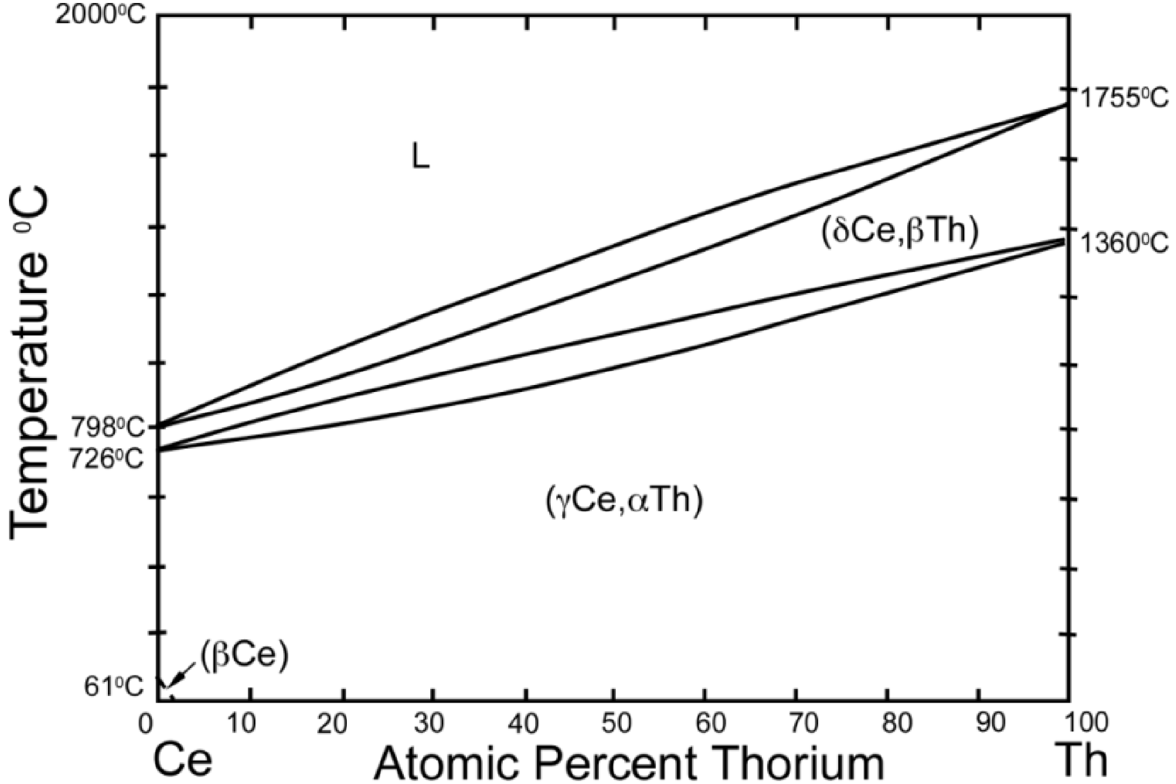
3 orders of magnitude difference (ε vs. δ)

Similar to liquid-solid

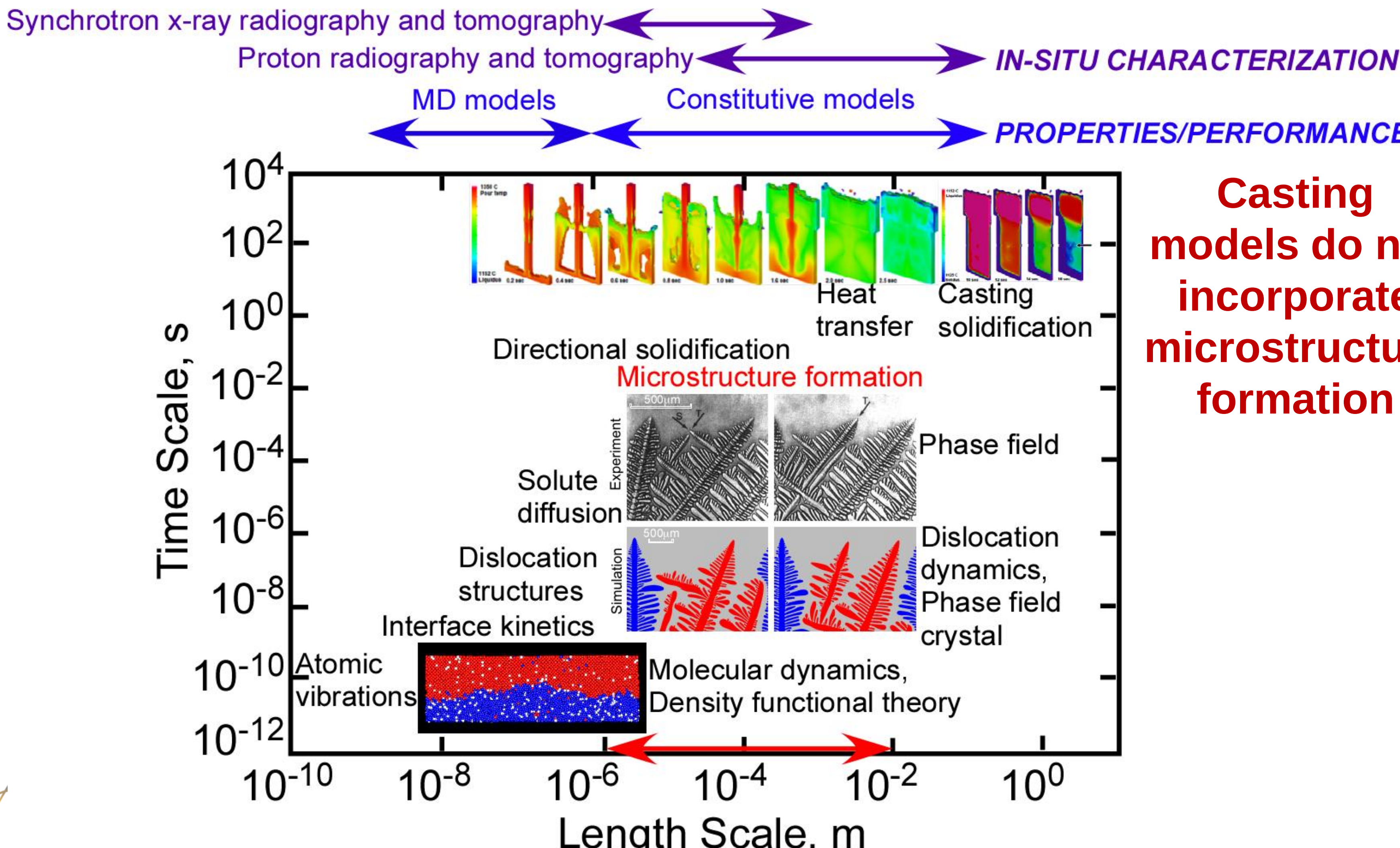
J.N. Mitchell et al., Metallurgical and Materials Transactions A, 2001, 32A:649-659

Phase Transformations in Ce-Th

- Segregation occurs during solidification
- Solid-state transformation in Th-rich regions at low T $\rightarrow$ large volume change



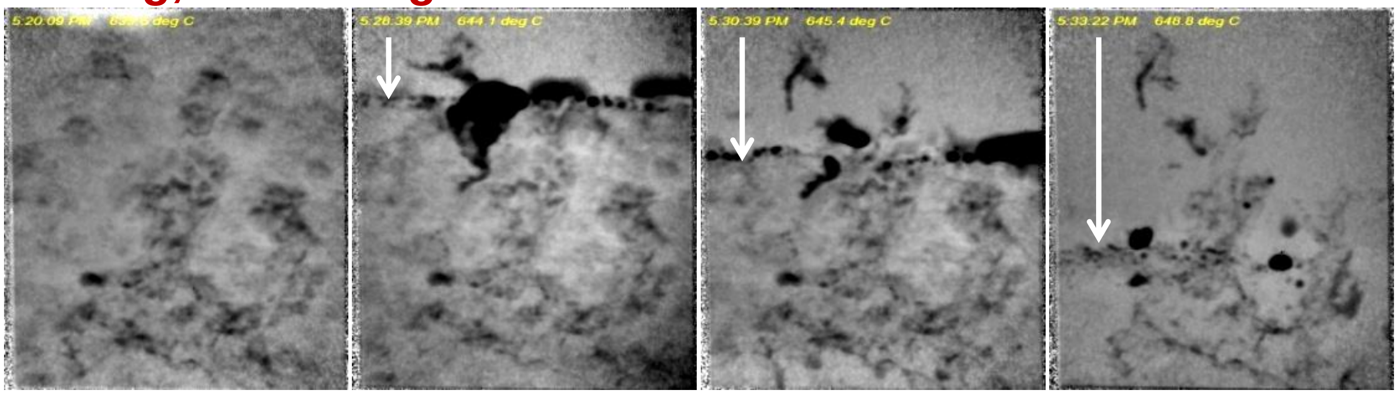
Solidification Phenomena at Various Length and Time Scales - Directed Synthesis and Processing to Control Microstructure



Casting models do not incorporate microstructure formation

In-situ Monitoring of Alloy Melt Fluid Flow and Solidification Using pRad at LANL (August 2011)

Melting, Increasing Time



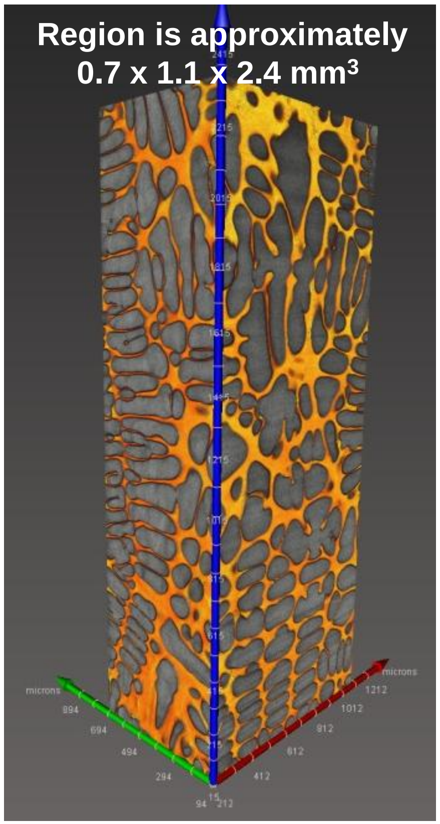
Solidification, Increasing Time

Al-In, X3 magnifier, > 1000 mm^2 field of view, 6 mm thick

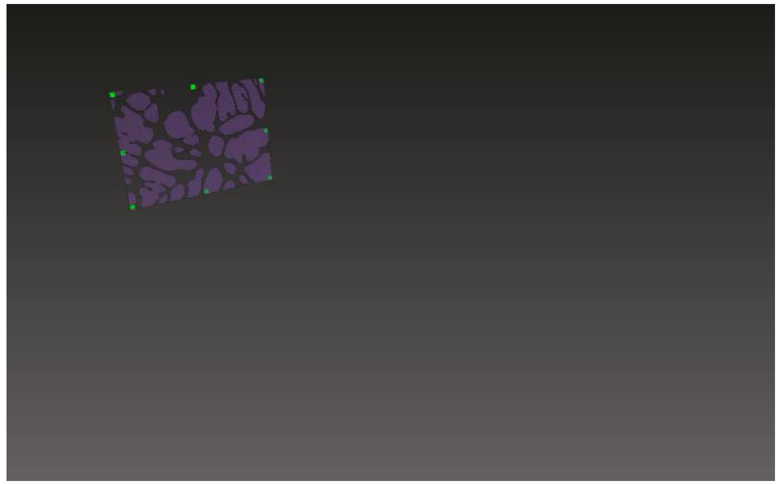
In-situ definition of solid-liquid interfaces and velocities, solute segregation, and alloy melt fluid flow

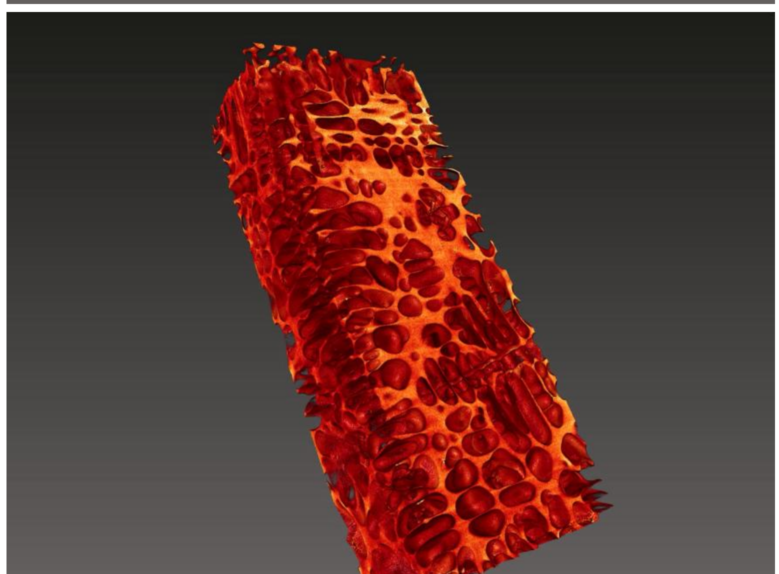
Early X-ray Radiography and Tomography at LANL - 3D Imaging of Materials on a Path to MaRIE (Real-Time)

Xradia micro-CT

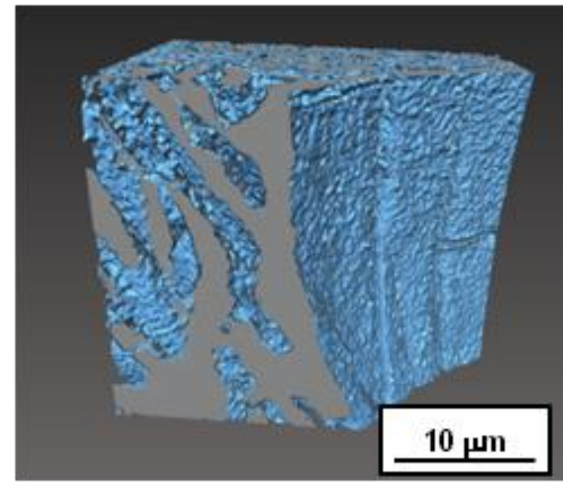


3 μm resolution in 3D





Xradia nano-CT



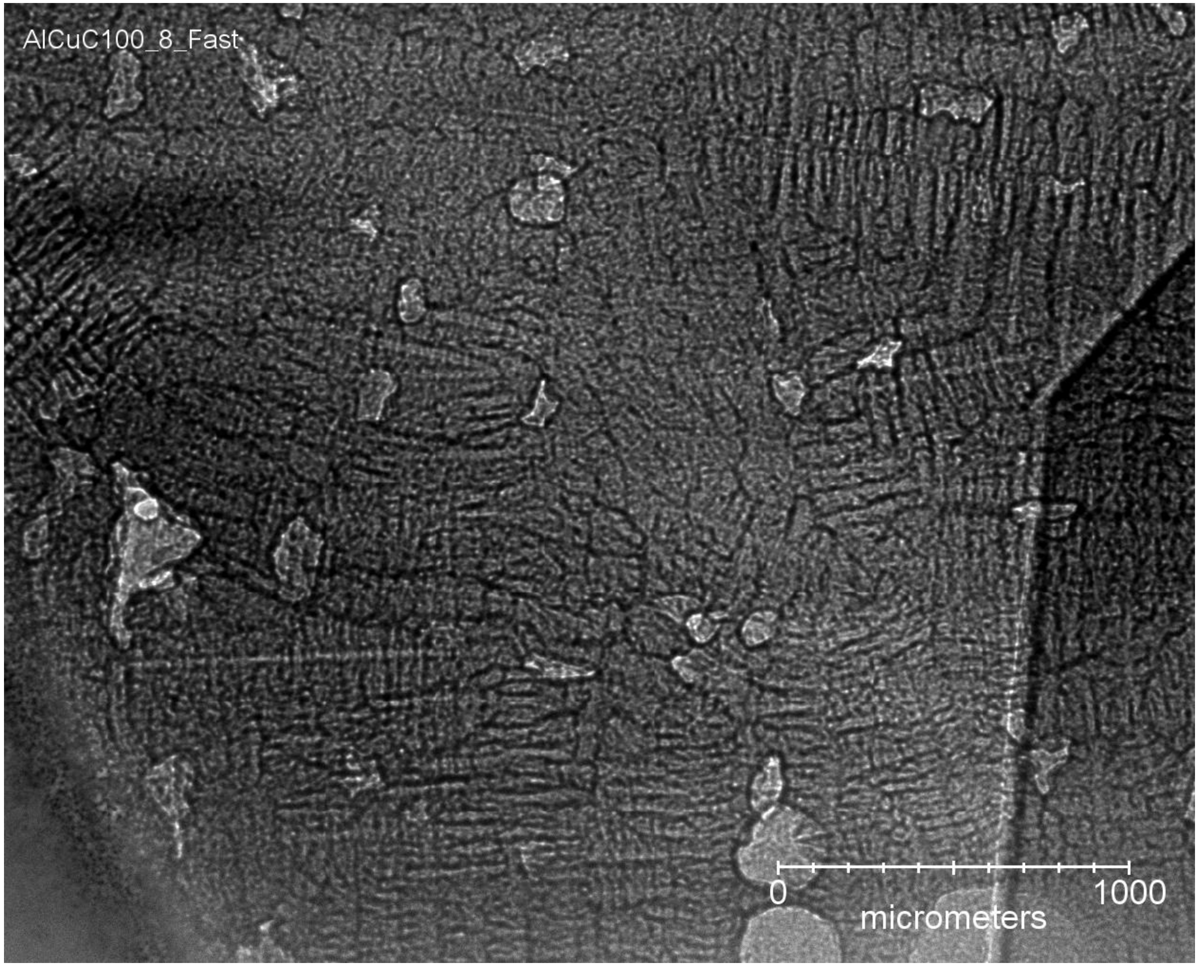
50 nm resolution in 3D

Resolving the two phase interdendritic region in Al-Cu

Synchrotron X-ray Radiography during Solidification at APS (December 2011); Al-Cu, Slow Continuous Cooling

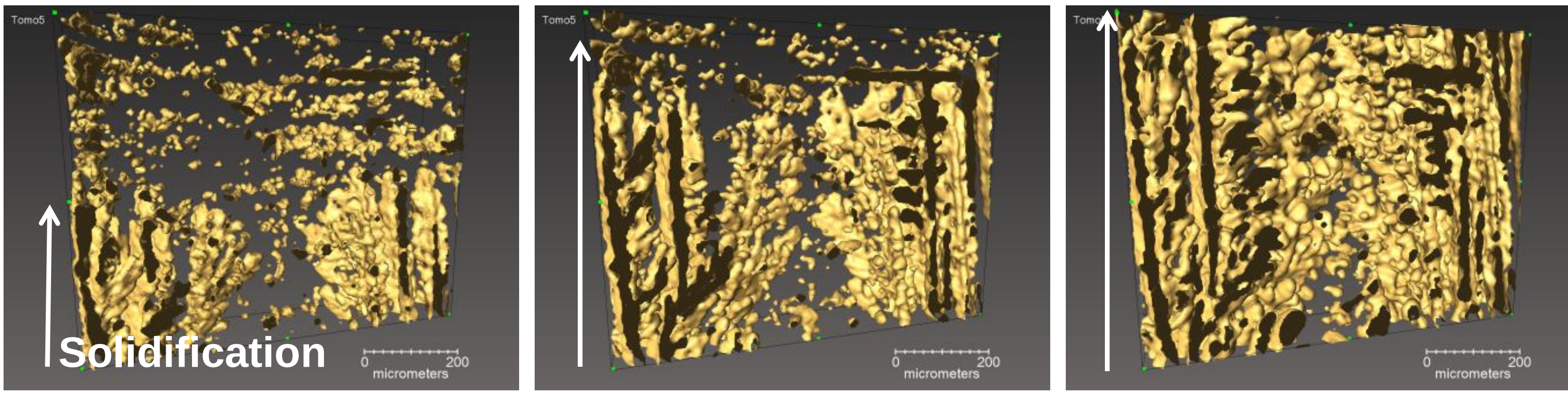
100 μm thick, < 10 mm^2 field of view

Synchrotron X-ray Radiography during Solidification at APS (December 2011); Al-Cu, Fast Continuous Cooling



100 μm thick, < 10 mm^2 field of view

3D Dendritic Growth and Coarsening during Solidification Using Synchrotron X-ray Tomography at APS (December 2011); Al-Cu



Increasing Time

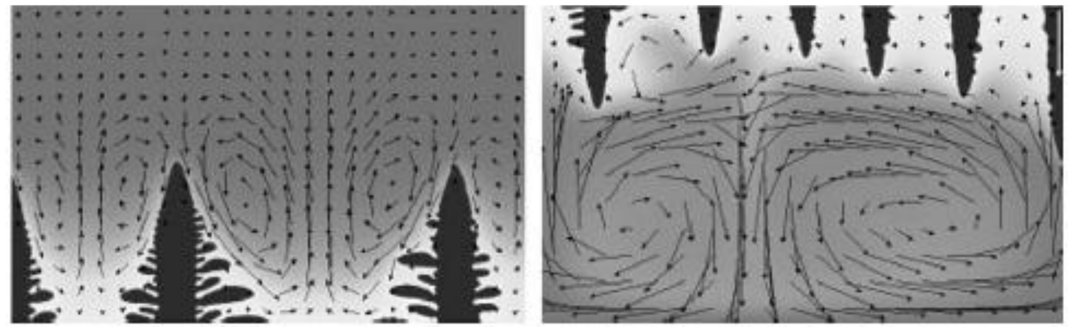
1 mm diameter cylindrical sample

Phase-Field Modeling

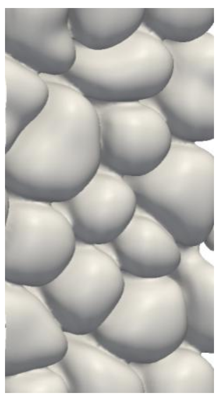
- Well-established for microstructure evolution
- Phenomenological, NOT ab-initio
  - Needs experiments to constrain parameters
- Best hope for multiscale simulations, integrating microstructure evolution with finite elements
  - Successfully demonstrated for solids (e.g., INL fuel performance codes)
  - Development needed for phase transformations

Predictive Mesoscale Phase-Field Models for Microstructure Evolution

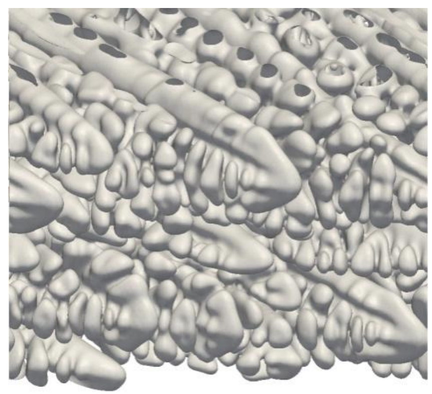
- 2D phase-field simulations
  - Thermo-solutal convection in Al-Cu
  - Solute varies from 4% (dark) to 7% Cu (light)



Equiaxed



Cellular



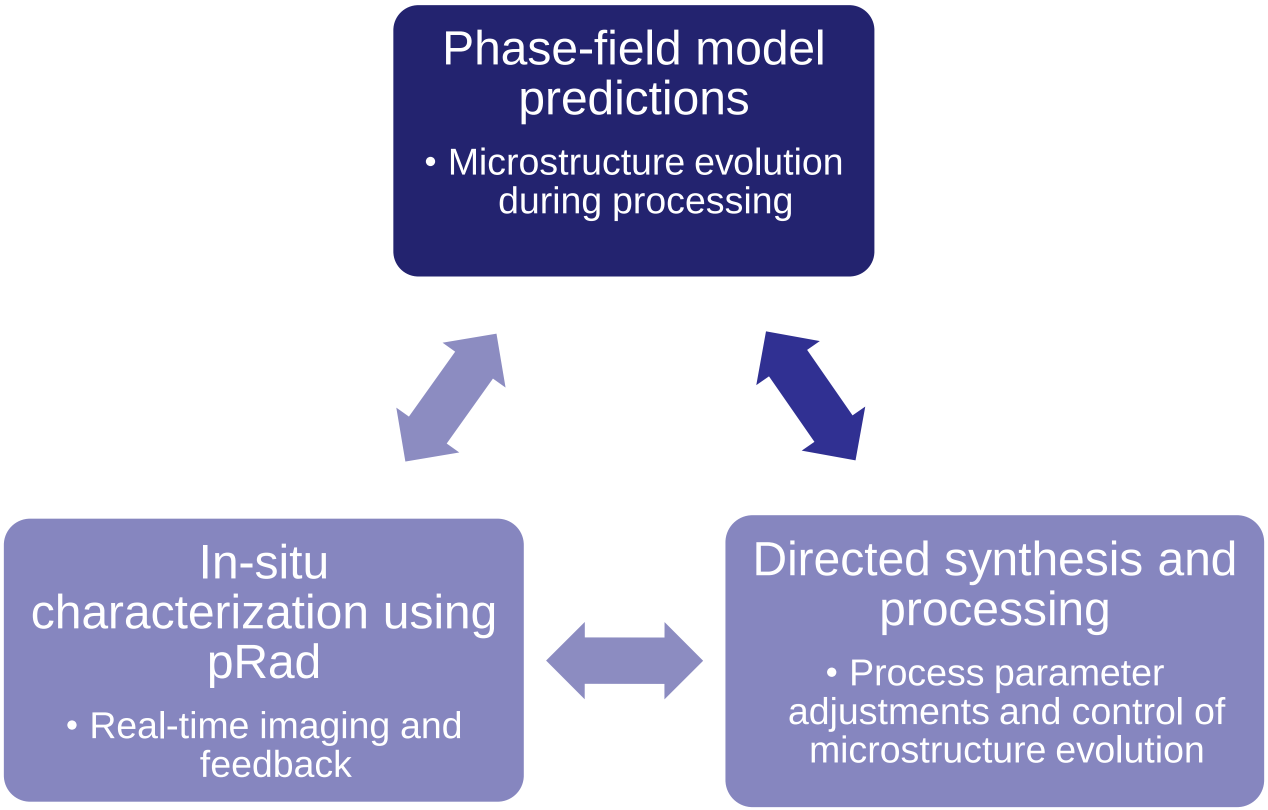
Dendritic

- 3D phase-field simulations (parallel implementation of the phase-field model)
  - Binary alloy microstructures
  - Different solidification processing parameters
- Expand to solid-state phase transformations for Pu-Ga

Same spatial and temporal scale as the experiments

Experiments ↔ Models

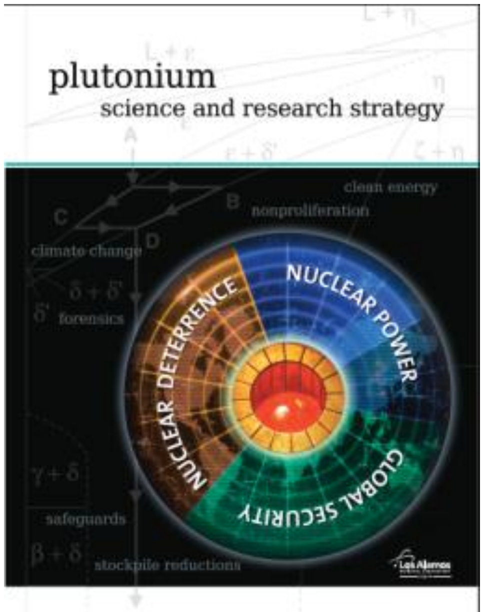
Control of Microstructure Evolution to Enable Co-design

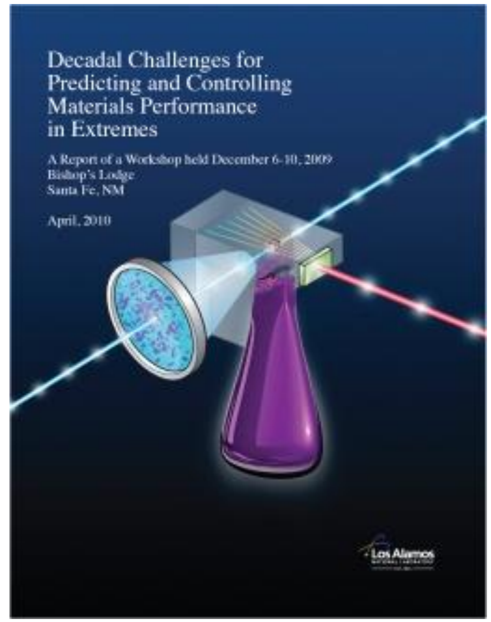


Definition of next-generation MaRIE capabilities required for process-aware manufacturing

Importance for the Laboratory

Materials: Discovery Science to Strategic Applications



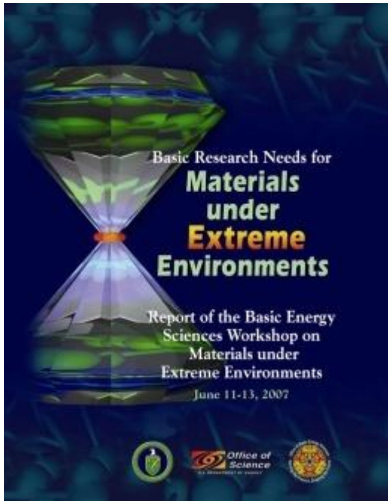


Plutonium strategy, materials in extremes, MaRIE vision

Recent workshops (dynamic experiments)

Phase transformations (P,T) and kinetics, materials processing and synthesis

Importance for the Nation

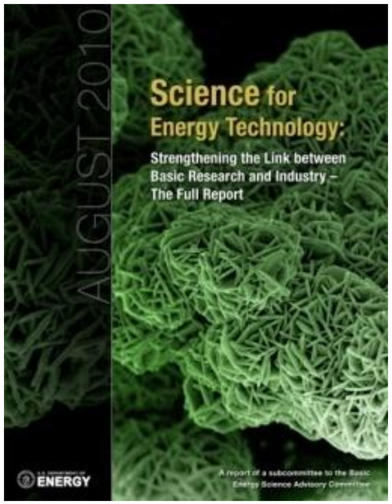


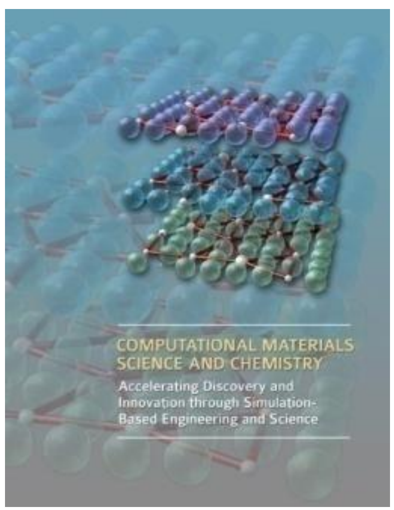
Basic Energy Sciences

Extreme environments, materials by design, ICME

Mesoscopic Materials and Chemistry (BES)

http://Meso2012.com



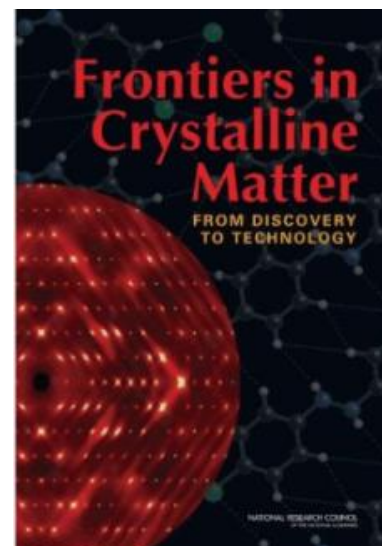


White House Office of Science and Technology Policy

Accelerating materials innovation from discovery to deployment; integrated approach

National Academies

Discovery and growth of crystalline materials; user facilities needed



Unprecedented Studies of Phase Transformations during Processing, Coupled with Theory and Modeling

- Major facility + advanced modeling + materials capability
  → Unique to Los Alamos
- Modeling non-linear instabilities is difficult
  → Los Alamos is good at this
- Crucial to national security materials work
- First full demonstration of the proposed value of MaRIE
  → Capabilities needed for process-aware manufacturing
- We are heading down this path
  → Advanced manufacturing, materials genome, BES mesoscale science initiatives…